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Citation for final published version:

Gill, R. T., Thornton, S. F., Harbottle, Michael John ORCID: <https://orcid.org/0000-0002-6443-5340> and Smith, J. W. N. 2016. Sustainability assessment of electrokinetic bioremediation compared with alternative remediation options for a petroleum release site. *Journal of Environmental Management* 184 (1) , pp. 120-131. 10.1016/j.jenvman.2016.07.036 file

Publishers page: <http://dx.doi.org/10.1016/j.jenvman.2016.07.036>
<<http://dx.doi.org/10.1016/j.jenvman.2016.07.036>>

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Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Sustainability assessment of electrokinetic bioremediation compared with alternative remediation options for a petroleum release site

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ARTICLE INFO

Article history:

Received 25 November 2015

Received in revised form

22 May 2016

Accepted 12 July 2016

Available online xxx

Keywords:

Sustainable remediation

Electrokinetic bioremediation

Green remediation

MTBE

ABSTRACT

Sustainable management practices can be applied to the remediation of contaminated land to maximise the economic, environmental and social benefits of the process. The Sustainable Remediation Forum UK (SuRF-UK) have developed a framework to support the implementation of sustainable practices within contaminated land management and decision making. This study applies the framework, including qualitative (Tier 1) and semi-quantitative (Tier 2) sustainability assessments, to a complex site where the principal contaminant source is unleaded gasoline, giving rise to a dissolved phase BTEX and MTBE plume. The pathway is groundwater migration through a chalk aquifer and the receptor is a water supply borehole. A hydraulic containment system (HCS) has been installed to manage the MTBE plume migration. The options considered to remediate the MTBE source include monitored natural attenuation (MNA), air sparging/soil vapour extraction (AS/SVE), pump and treat (PT) and electrokinetic-enhanced bioremediation (EK-BIO). A sustainability indicator set from the SuRF-UK framework, including priority indicator categories selected during a stakeholder engagement workshop, was used to frame the assessments. At Tier 1 the options are ranked based on qualitative supporting information, whereas in Tier 2 a multi-criteria analysis is applied. Furthermore, the multi-criteria analysis was refined for scenarios where photovoltaics (PVs) are included and amendments are excluded from the EK-BIO option. Overall, the analysis identified AS/SVE and EK-BIO as more sustainable remediation options at this site than either PT or MNA. The wider implications of this study include: (1) an appraisal of the management decision from each Tier of the assessment with the aim to highlight areas for time and cost savings for similar assessments in the future; (2) the observation that EK-BIO performed well against key indicator categories compared to the other intensive treatments; and (3) introducing methods to improve the sustainability of the EK-BIO treatment design (such as PVs) did not have a significant effect in this instance.

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1. Introduction

The management of contaminated land is a global challenge. Its restoration is often considered to provide net positive benefits, but if remediation practices are selected and implemented poorly more environmental impact can arise than is associated with the contamination. Integrating sustainability practices into contaminated land remediation provides an opportunity for social, environmental and economic benefits of the process to be considered and optimised. Sustainable remediation is defined by the

Sustainable Remediation Forum, UK (SuRF-UK) as “the practice of demonstrating, in terms of environmental, economic and social indicators, that the benefit of undertaking remediation is greater than its impact” (CL:AIRE, 2010). There are two ways in which sustainable remediation can be applied at contaminated sites (NICOLE, 2010): 1) at the management level, integrating sustainability assessments into the wider decision making process; and 2) at the site-specific level, by an assessment to compare options against certain sustainability indicators. SuRF-UK has produced a framework which provides a structure for implementing these two approaches within a contaminated site project. The framework has two stages: Stage A, plan and project design; and Stage B, remediation option appraisal and implementation. This study applied

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Stage B of the framework, by applying a sustainability assessment to contaminated site remediation technology selection.

The SuRF-UK sustainable remediation framework describes a tiered approach to sustainability assessments. There are three tiers, each requiring increasing amount of data for the assessment: Tier 1 is qualitative (e.g. simple rankings against ideal criteria); Tier 2 is semi-quantitative (e.g. multi-criteria analysis); and Tier 3 is quantitative (e.g. cost-benefit analysis). The steps associated with an assessment include (Bardos, 2014; Bardos et al., 2011): 1) defining remediation objectives to identify the decision that is being supported; 2) stakeholder engagement; 3) identifying boundaries of the assessment such as system, lifecycle, spatial and temporal; 4) identifying relevant sustainability indicators for the scope of the assessment; 5) defining the assessment methodology, i.e. either Tier 1, 2 or 3 or a combination; 6) conducting the sustainability assessment and 7) verifying and reporting the results.

Several case studies apply the SuRF-UK framework to contaminated sites and demonstrate the economic, environmental and social benefits of the process. For example, a Tier 1 assessment was applied to a fuel storage depot in Madeira, Portugal, concluding that enhanced bioremediation to be a more sustainable approach than thermal desorption, based largely on reduced cost and CO₂ emissions, but with an associated longer duration for remediation activity (CL:AIRE, 2013a). Additionally, Tier 2 and 3 assessments were completed at a former airbase site where aviation fuel was thought likely to impact a primary aquifer. It concluded that environmental and social impacts out-weighed the economic, resulting in a more expensive but more sustainable and operationally better solution (CL:AIRE, 2013b).

A novel aspect of this study is the inclusion of electrokinetic-enhanced bioremediation (EK-BIO) within risk management. Electrokinetics is the application of a direct current to the subsurface to initiate solute transport independent of hydraulic conductivity, by electroosmosis, electromigration and electrophoresis (Acar and Alshawabkeh, 1993). These transport processes can be used to enhance bioremediation at a range of scales (Gill et al., 2014). At the micro-scale, this can help increase bioavailability and bio-accessibility (Wick et al., 2007). At the macro-scale, electron acceptors and/or nutrients can be delivered into the contaminated zone to support biodegradation (Lohner et al., 2008). Furthermore, these transport processes can be as effective in heterogeneous sediments with significant hydraulic conductivity contrasts (Gill et al., 2015). The technology is considered a good candidate for sustainable remediation as the principal costs after set up are electricity and the amendment used (Alshawabkeh et al., 1999; Kim et al., 2014). Consequently, there is significant interest in coupling electrokinetics with other remediation technologies and incorporating it as part of remediation options appraisal will further advance the state of knowledge.

The aim of the study was to assess the sustainability of different remediation options, including the theoretical application of EK-BIO, for a gasoline/MTBE contaminated site. The objectives were to:

1. Perform Tier 1 and Tier 2 sustainability assessments on a site contaminated by an unleaded gasoline release from a petrol filling station and use the findings to inform a management decision;
2. Include EK-BIO in the remediation option appraisal, using an electron balance model to inform operational parameters such as treatment duration, power (electricity) consumption and amendment usage; and
3. Investigate the effect of incorporating photovoltaics and limiting amendment usage on the EK-BIO remediation option using different scenarios relative the base case above.

Currently there are no reported examples of using electrokinetic bioremediation within a sustainability assessment, or how modifications to the treatment design, such as inclusion of photovoltaics, influence the overall sustainability performance. These are important knowledge gaps in the development of electrokinetic remediation. Furthermore, this is the first peer-reviewed application of the SuRF-UK framework.

2. Conceptual site model

The focus of this study is a petrol filling station (PFS) site located up hydraulic gradient of a water supply well (WSW). There was a fuel release into the subsurface at the PFS resulting in the fuel additive methyl tert butyl ether (MTBE) detection in the WSW. The PFS was decommissioned, the fuel release stopped, and investigation and remediation undertaken. Several groundwater sampling and monitoring events have been completed at the site to assess the risk posed by MTBE to the WSW. Remedial action to date includes the installation of a hydraulic containment system (HCS) to break the source-pathway-receptor (SPR) linkage, and soil vapour extraction (SVE) and multi-phase extraction (MPE) to treat mobile and residual-phase LNAPL near the source zone.

2.1. Site geology and hydrogeology

The main hydrogeological units in the shallow subsurface at the site are summarised in Table 1 and a cross section in Fig. 1A. The top of the Cretaceous Chalk aquifer is located at around 20 m BGL, and forms a regionally important water supply aquifer. The Chalk is overlain by ca. 20 m low permeability clay till, through which a glacial sand channel was cut. The channel sands are a mix of high permeability sands and gravels interspersed with low permeability silt lenses. Regional groundwater flow is towards the north east, however, the local hydrogeological regime is modified by abstraction at the WSW, which draws Chalk groundwater in an easterly direction. When the WSW is on, groundwater flow in the channel sands and chalk is towards the well creating a downward vertical hydraulic gradient in the channel sands. When the WSW is not pumping the regional groundwater flow is dominant and the hydraulic gradient between the channel sands and chalk aquifer is reversed. The water table fluctuates under the influence of the abstraction and seasonal variations.

2.2. Contaminants of potential concern

Numerous petroleum hydrocarbon constituents are present on site. Those exceeding UK drinking water standard or World Health Organisation appearance taste and odour values at the highest number of locations include benzene, toluene, ethyl-benzene, xylene (BTEX) and MTBE. These compounds are considered the main contaminants of potential concern, consistent with other gasoline impacted sites (Bowers and Smith, 2014). Hydrocarbons were present in both free phase and dissolved phases. The free-phase has migrated south-east into the channel sands, with significant smearing due to groundwater fluctuation. The dissolved-phase within the channel sand is drawn down by the vertical

Table 1
Summary table of the geological units present on site.

Geological unit	Hydraulic conductivities (m s ⁻¹)
Channel Sands	1.5×10^{-5} to 1.2×10^{-9}
Glacial till	1.2×10^{-8} to 1.2×10^{-12}
Chalk	1.2×10^{-3} to 3.5×10^{-4}

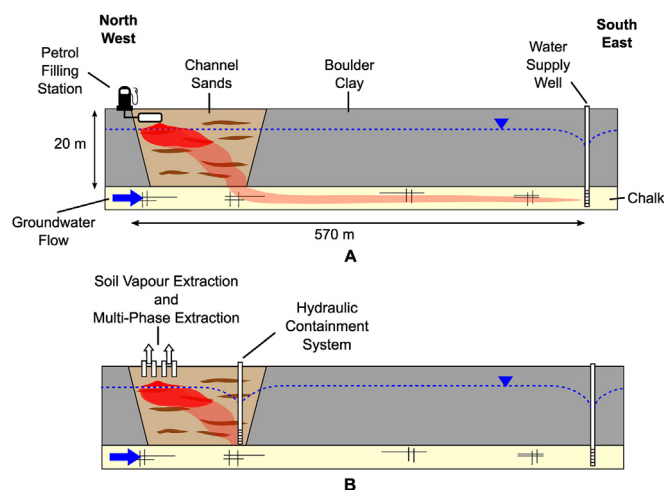


Fig. 1. Cross section of field site split into two parts, A and B. Conceptual site model showing A. geological and hydrogeological features and the principal source – pathway – receptor linkage present on site; and B. cross section following the first stage of remedial action.

hydraulic gradient towards the chalk aquifer (Fig. 1A) and the WSW, 570 m down gradient. The highest observed concentrations for contaminants of concern are 6 m below groundwater level at: benzene, 52.6; toluene, 63.2; xylene, 18.9; ethylbenzene, 2.8; and MTBE, 23.5 (values in mg L^{-1}).

The distribution of dissolved-phase organic contaminants is influenced by the availability of electron acceptors supporting biodegradation. BTEX and MTBE are present within the channel sands and up to the boundary with the Chalk aquifer. Within the channel sands, BTEX are biodegraded using dissolved oxygen, nitrate, sulfate, and mineral-derived Fe and Mn oxides, based on the groundwater quality data. Biodegradation of MTBE occurs most efficiently under aerobic conditions (Shah et al., 2009), although anaerobic biodegradation has been reported (Somsamak et al., 2001, 2005; and 2006) it cannot be deduced in the groundwater quality data. Thus the excess of BTEX, which degrades more readily than MTBE under aerobic conditions (Wiedemeier et al., 1999), creates competition for dissolved oxygen. Only MTBE is present in the Chalk aquifer and is biodegraded aerobically, based on observed elevated concentrations of its primary biodegradation product, *tert*-butyl alcohol (TBA) (Shah et al., 2009), although not at a rate sufficient to mitigate risk. It is anticipated that if the mass of BTEX is removed or reduced from the channel sands this will reduce the BTEX mass flux into the Chalk aquifer and therefore increase the aerobic biodegradation of MTBE.

Based on these observations the principal SPR linkage that drives risk management requirements on site is related to MTBE migration (Table 2). Remedial targets for the site consider: 1) MTBE biodegradation rates within the Chalk aquifer; 2) dilution of MTBE at the WSW and during migration between the channel sands and Chalk aquifer; and 3) an agreed remedial objective for MTBE that prevents taste and odour impact at the abstraction well.

Two remedial actions have been implemented to manage the

risk of MTBE to the WSW (Fig. 1B). Firstly, a HCS installed at the boundary between the channel sands and Chalk aquifer to continually extract contaminated water for discharge and treatment. This breaks the SPR linkage, by preventing MTBE from entering the Chalk aquifer, and has been validated by frequent monitoring events over the four years since installation. Its continued operation is critical to mitigate risk to the WSW. Secondly, SVE and MPE systems have been installed to remove hydrocarbon mass from the source zone. The sustainability assessment focuses on determining which techniques are appropriate to treat the remaining residual NAPL and dissolved phase contaminants in the source areas. The rationale is to reduce the magnitude and duration of the contaminant flux into the Chalk aquifer and therefore reduce the duration the HCS is active.

2.3. Identified remediation options

The zone of contamination covers a surface area of 1500 m^2 ($75 \text{ m} \times 20 \text{ m}$) and extends 6 m below the water table (ca. 7.5 mbgl). Four *in situ* remediation technologies have been identified for appraisal, all run concurrently with the HCS:

1. **Monitored Natural Attenuation (MNA)** – MNA is the least intensive technique. It is justified on this site as biodegradation of the BTEX and MTBE is observed in the channel sands aquifer and HCS provides protection to the abstraction well. Treatment design includes utilising existing monitoring wells, with four monitoring events in the first year, followed by one in each subsequent year of operation, adopting a lines-of-evidence approach in line with good practice regulatory guidance (EA, 2000)
2. **Electrokinetic Enhanced Bioremediation (EK-BIO)** – The EK-BIO technique is suitable for application to this site due to the physical heterogeneity over the specified range of hydraulic conductivities in the contaminated channel sands aquifer (Gill et al., 2015). The aim is to introduce nitrate to supplement the global supply of electron acceptors and directly address the limitation on anaerobic BTEX degradation. This will restrict the mass and dimensions of the BTEX plume and reduce the competition with MTBE for electron acceptors leading to lower mass flux of MTBE into the Chalk aquifer. Electrodes are arranged in a bidirectional configuration with a line of cathodes in the centre between two rows of anodes (see Fig. S1, and S2 supporting information) (Gill et al., 2014). The treatment will be conducted in three phases to accommodate optimal electrode distances. A constant voltage gradient of 50 V m^{-1} is assumed with a drop of 60% in the zone adjacent to the cathode due to the increase in electrical conductivity from the amendment (Wu et al., 2012). Subsequently, the voltage gradient drop will control the amendment flux into the system.
3. **Air Sparge/Soil Vapour Extraction (AS/SVE)** – This technique is suitable because of the existing SVE infrastructure and pilot-scale testing of the AS system indicate the treatment could be effective for treating the high-K zones on site. The treatment will be conducted in two phases to accommodate the short AS well radius of influence (ca. 3.3 m) and subsequent high number

Table 2

The principal SPR linkage present on site and remedial target for MTBE as agreed with the regulator.

Source	Pathway	Receptor	Remedial targets
Dissolved and free-phase MTBE	Dissolution and migration through the saturated channel sand and Chalk aquifer.	Taste and odour impacts resulting from consumption of water extracted from the well.	MTBE: 3.3 mg L^{-1} (at source zone, based on achieving taste and odour threshold at point of abstraction)

of treatment wells. The treatment module associated with the AS/SVE system is a catalytic oxidation unit.

4. *Pump and Treat (PT)* – This technique is suitable due to the relatively high hydraulic conductivity of the channel sands and Chalk aquifers. In addition, MTBE is more soluble than other contaminants, with limited retardation by sorption to the aquifer matrix. Two pumping wells with a radius of influence of 20 m are sufficient to cover the treatment area, assuming a 5% drawdown of the water column and hydraulic conductivity of $1.5 \times 10^{-5} \text{ m s}^{-1}$ consistent with the most permeable sections of the channel sands. The treatment modules associated with the PT system comprise an oil/water separator, air stripper and granular activated carbon unit.

Realistic treatment durations for each technique have been assigned using sources from site-specific reports and literature values. These include minimum, medium and maximum values (Table 3). A simple electron balance model (EBM) was developed (similar to the one in Gill et al., 2014) to inform EK-BIO treatment durations and other operational parameters as currently no field data exist in the literature for its application to dissolved phase LNAPL contaminants. An EBM determines the length of time required for the number of electron acceptors to equal electron donors (Thornton et al., 2001), in this example, the nitrate amendment and BTEX and MTBE contaminants represent the electron acceptors and electron donors respectively. The number of electrons accepted or donated depends on the stoichiometry of each half reaction (see Table S1 in supporting information). One-dimensional electromigration mass flux equations were applied to simulate nitrate transport into the treatment domain until a sufficient amount had been added to equalise the electron donor mass (see Table S2 and S3 supporting information) (Acar and Alshawabkeh, 1993). This duration was then added to the length of time required for nitrate to migrate through the treatment domain (Alshawabkeh et al., 1999). The treatment domain was split into three layers to represent the heterogeneity observed on site, material properties were taken from Gill et al. (2015). In the electron balance model BTEX in the channel sands aquifer is assumed to be biodegraded anaerobically using nitrate (Wiedemeier et al., 1999). Anaerobic respiration of MTBE using nitrate has been demonstrated (Bradley et al., 1999, 2001), but cannot be proven at the site from the groundwater quality data. Instead, MTBE is assumed to be aerobically biodegraded to TBA in the Chalk aquifer (Spence et al., 2005). In this way, the EK-BIO treatment is used to enhance the nitrate flux for BTEX biodegradation, allowing aerobic respiration of MTBE further down-gradient in the sand and Chalk aquifers.

The minimum, medium and maximum duration ranges were calculated using a range of sources. For MNA a range of durations is taken from site specific modelling reports; for EK-BIO the transport properties of the materials simulated in the EBM were varied according to literature values (Table S3, supporting information) (Gill et al., 2015); for AS/SVE a range of volatile organic carbon extraction rates were taken from pilot trials (Table S4 and S5 supporting

information); and PT a range of attenuation rates from MTBE contaminated sites were used (Table S6 and S7 supporting information) (McHugh et al., 2014). Further details of treatment design specifications and associated assumptions are given in the supporting information.

3. Sustainability assessment framework

3.1. Remediation objectives and stakeholder engagement

The sustainability assessment covers Stage B of the SuRF-UK framework, with the overall task of selecting the most sustainable remedial option to deliver project objectives (CL:AIRE, 2010). The remedial objectives are: 1) to achieve risk-based close out criteria for MTBE in groundwater; and 2) return properties adjacent to the source area back into beneficial use. The stakeholders in this project included the local authority, Environment Agency, water abstraction owner, the site owner and their professional advisors.

3.2. Assessment boundaries

This sustainability assessment is constrained by four types of boundary conditions. Firstly, system boundaries; the processes associated with remedial operations to achieve the risk management objectives. An example from the PT option includes establishing site infrastructure, drilling pumping wells, extracting groundwater etc. System boundaries are shown in supporting information, Fig. S3 as solid boxes. Secondly, life-cycle boundaries; the materials and energy inputs required for a step in the remediation process, as well as the outputs from that step, such as air emissions from transport or remediation activity. The analysis excludes manufacture of remediation equipment; it is assumed to be rented from a supplier or purchased with the aim of future use. The lifecycles associated with the four options are shown in Fig. S3 as dashed boxes. Inputs and outputs are shown for each step in the technique. Thirdly, spatial boundaries extend to the area around the site, with the footprint of the dissolved phase plume and transport to and from site. Fourthly, temporal boundaries exist as long as the pollutant linkages and risk management options are required, or as long as the dissolved phase plume in the channel sands aquifer exists. This is shown in Fig. S3 as the diamond-shaped decision box. Some aspects of remediation techniques are not included in the overall analysis as they are assumed to be in place at the start of the remediation option appraisal. These include drilling monitoring boreholes and establishing site infrastructure and drilling treatment boreholes for the SVE and HCS systems.

3.3. Scope of assessment

The sustainability assessment covers the fifteen indicator categories (ICs) described in SuRF-UK documentation across the three 'pillars' of sustainability (CL:AIRE, 2011). The purpose of the indicator set is to make the sustainability assessment more transparent

Table 3
Summary of treatment durations and calculation method used to inform the sustainability assessment.

Option number	Remediation option	Treatment duration range (years)			Calculation method
		Min	Med	Max	
1	MNA + HCS	15	20	25	Site reports and modelling study
2	EK-BIO + HCS	3.4	6.0	7.3	Electron balance model
3	AS/SVE + HCS	3.8	5.0	7.5	Site reports from pilot test
4	PT + HCS	5.2	6.4	7.0	Literature data based on multiple active MTBE sites (McHugh et al., 2014)

to assessors and stakeholders, as well as facilitating both a Tier 1 and Tier 2 assessment. All fifteen ICs are included to provide the most holistic view. ICs considered to be a priority were identified by consultation with stakeholders, who selected two priority ICs from each pillar. The two ICs from each pillar with the most number of priority selections are highlighted with an asterisk (*) (Table 4). The semi-quantitative Tier 2 assessment required the identification of quantifiable factors within each IC, subject to data availability. Where no additional factors could be identified the ranking from the qualitative Tier 1 assessment was carried over to the Tier 2 assessment. Care was taken not to replicate scores between ICs. For some ICs more than one metric could be identified, for example, natural resources and waste includes four different metrics: water discharge from treatment, volume of soil material displaced, raw materials used for well construction and volume of fuel consumed. The groundwater quality IC includes quantitative and qualitative metrics. The quantitative metric is the value of groundwater in the channel sands lost due to abstraction at the HCS that otherwise would have been abstracted at the WSW (Bartlett et al., 2014) and the qualitative metric that considers the broader impacts on groundwater quality, such as groundwater chemistry that are harder to quantify.

3.4. Assessment approach

The qualitative Tier 1 assessment comprised a simple ranked comparison of the different remediation options using generic and conservative assumptions against defined sustainability ICs. The semi-quantitative Tier 2 assessment applied a multi-criteria analysis (MCA) using site-specific data to the same sustainability ICs. For the Tier 1 analysis the middle treatment durations from Table 3 were used for all treatments. In Tier 2, treatment durations were subject to an uncertainty analysis that applied the minimum and maximum estimated treatment durations shown in Table 3.

4. Tier 1 sustainability assessment

4.1. Tier 1 methodology

A ranked score between 1 and 4 was assigned for the different options framed against an idealised scenario. For example, air emissions (ENV 1*) the ideal scenario is no air emissions and direct

economic cost (ECON 1*) the ideal scenario is minimal capital, operational and management cost (see Table S8 supporting information for descriptions of ideal scenarios for other ICs). A higher rank (i.e., 4) was assigned to the least sustainable option and a low rank (i.e., 1) to the most sustainable option. At Tier 1 all ICs were weighted equally, i.e. a priority IC such as air emissions had the same weight as a non-priority IC such as ecology. Qualitative ranking was supported by evidence from different categories and includes qualitative and basic quantitative assessment (see Table S9 – S11 supporting information). Equal rankings were allowed if differences between options were <10%. The treatment durations for the assessment were the middle estimates as defined above. Uncertainty regarding treatment durations was reduced by comparing the individual ICs against an ideal scenario. It was also included as part of the social category of ICs.

4.2. Tier 1 results

The rankings and justifications for individual ICs are given in Table S12 supporting information. The cumulative rankings of the different options are shown against all ICs and priority ICs in Fig. S4, supporting information. Option 3 (AS/SVE+HCS) appears to be the best option when all ICs are considered. This is due to: 1) shorter treatment duration of ICs that are time-dependent, such as indirect or induced economic benefits; 2) added certainty on treatment performance from pilot-scale study; and 3) a function of the AS/SVE system used to treat both unsaturated strata and groundwater, thus improving soil and groundwater conditions. However, for priority ICs identified during the stakeholder workshop Option 2 (EK-BIO+HCS) and Option 3 are equivalent. This is due to Option 2 scoring well against priority ICs, for example, air emissions and total direct economic cost are lower than Option 3 and Option 4 (PT+HCS) because no treatment plant is used. However, against non-priority ICs Option 2 is less favorable because of high levels of uncertainty compared with other Options and relatively long treatment duration compared to Option 3.

The Tier 1 assessment identifies a choice of two management decisions. Firstly, select Option 3 for the site as it is the most sustainable when all ICs are considered and hence is justified at this level. Secondly that the assessment should progress to Tier 2 and attempt to further differentiate between Options especially against priority ICs. For the purposes of this study the second option will be applied.

Table 4

Indicator set with identified priority ICs. ICs used in the Tier 1 qualitative assessment and Tier 2 semi-quantitative assessment are shown.

Criteria	Indicator category	Label	Priority	Tier 2 metric	Unit
Environment	Emissions to air	ENV 1	*	Total CO ₂ emissions	kg CO ₂ -e
	Soil and ground conditions	ENV 2		Qualitative (Tier 1)	n/a
	Groundwater quality	ENV 3	*	Value of water lost to HCS extraction	GBP (£)
	Ecology	ENV 4		Qualitative (Tier 1)	n/a
	Natural resources and waste	ENV 5		Qualitative (Tier 1)	n/a
Economic				Water discharge from remediation treatment only	m ³
				Volume displaced soil material	m ³
				Raw materials used in well construction	kg PVC
				Volume of petrol consumed	m ³
	Direct economic costs	ECON 1	*	Total economic cost	GBP (£)
	Indirect economic costs and benefits	ECON 2		Net present value of housing on site	GBP (£)
	Employment and employment capacity	ECON 3		Qualitative (Tier 1)	n/a
Social	Induced economic costs and benefits	ECON 4		Net present value of petrol filling station	GBP (£)
	Project lifespan and flexibility	ECON 5	*	Duration of treatment	Years
	Human health and safety	SOC 1	*	Time lost due to injury from operation	Hours
				Time lost due to injury from traffic accidents	Hours
	Ethics and equality	SOC 2		Qualitative (Tier 1)	n/a
	Neighborhoods and locality	SOC 3	*	Qualitative (Tier 1)	n/a
	Communities and community involvement	SOC 4		Qualitative (Tier 1)	n/a
	Uncertainty and evidence	SOC 5		Qualitative (Tier 1)	n/a

5. Tier 2 sustainability assessment

5.1. Tier 2 scoring method

Both quantitative and qualitative data sources for individual sustainability ICs were used to inform the MCA (Table 4). Further details on how the different metrics were calculated are provided in Table S13–S15 supporting information. The method of scoring the MCA is similar to that described by Postle et al. (1999):

$$\text{MCA Score} = \left(\frac{\text{Input Value for Option}}{\text{Maximum Value Across Options}} \right) \times 100 \times \text{Priority Weighting} \quad (1)$$

Numerical values for a particular IC were normalised against the maximum value across all treatment options and multiplied to provide a score between 0 and 100. An IC weighting was then assigned based on stakeholder priorities (Table 4). Where more than one metric is used an additional normalisation factor was applied to ensure that each metric contributed an equal amount to the overall IC score identified. For example, natural resources and waste, ENV 5, there are 4 metrics (Table 4) that each contribute 25% to the total IC score. As with the Tier 1 assessment, the higher the MCA score the lower the sustainability of the option.

MCA scoring methods for contaminated land options appraisal in the literature include similar components, namely: data for criteria and sub-criteria, a normalisation factor for criteria and sub-criteria and weightings for priority ICs. They differ in how the scores are calculated or presented. For example Harbottle et al. (2008) applies the Postle et al. (1999) method and consider the positive effects of remediation. In the present study, aftereffects of remediation are not included as it is assumed the benefits of remediation will be the same for all options. Furthermore, Blanc et al. (2004) develop a scoring method that produces a 'best' and 'worst' ranking of different technologies for a site.

Parameters used to derive the MCA scores are included in Table S16–S18 supporting information. They include the quantitative input values, IC and metric weightings and the final weighted MCA score. The same treatment designs are used as for the Tier 1 assessment. In addition, extra scores are compiled for three scenarios that examine the effect of sustainability enhancements applied to EK-BIO treatment as part of Option 2. These include Scenario 1: a photovoltaic array that provides electricity; Scenario 2: constant flushing of electron acceptors in uncontaminated groundwater through the electrode chambers, as opposed to the addition of amendment; and Scenario 3: a combination of scenario 1 and 2. These scenarios were not applied to AS/SVE or PT because their treatment design is unsuitable for the enhancements. For example, both include treatment plants that require a regular power supply to ensure treatment efficiency, and neither have an amendment with a low or no-cost substitute. EK-BIO is suitable because effective treatment has been demonstrated with variable amendment fluxes into sediment material that would arise from intermittent power supply (Mao et al., 2012; Wu et al., 2007). These scenarios assume that the HCS is active during treatment.

5.2. Tier 2 uncertainty analysis

For the Tier 2 assessment uncertainty is represented at the qualitative and quantitative level. Qualitatively uncertainty is represented by the IC, uncertainty and evidence (SOC 5) that

reflects confidence in treatment effectiveness based on quality of available evidence, similar to Tier 1. Quantitatively, the range of duration values shown in Table 3 were used to inform other time dependent metrics, i.e. all quantitative metrics with the exception of those specific to site setup (e.g. well drilling), qualitative rankings did not vary. Hence, quantitative uncertainty is based on variability in field conditions only and assumes each technique is effective.

5.3. Tier 2 sensitivity analysis

A sensitivity analysis was conducted to provide insight into which sustainability ICs had the greatest impact on the MCA score for a particular option (Rosén et al., 2015). The analysis requires a Monte Carlo simulation to be performed. This is a stochastic simulation where user defined ranges of uncertainty and probability distribution for different parameters are inputs that are propagated through the model. The output is a forecast that is based on the simulation being run numerous times. The sensitivity analysis is calculated by rank correlation of parameter inputs (independent variable) and the forecast outputs (dependent variable) then performing linear regression on ranked sets. Correlation coefficients are then tallied for each forecast and normalised (McNab and Doohar, 1998).

For this study minimum and maximum values for the different sustainability ICs derived from the uncertainty analysis were used to inform the input range for the simulation. A uniform probability distribution was assumed for all ICs. The output forecast was a range of MCA scores, the simulation was run 10,000 times using Monte Carlo simulation software, Crystal Ball for Microsoft Excel.

5.4. Tier 2 results

5.4.1. Economic indicator categories

The MCA scores for economic ICs showed no significant difference between options when the full minimum and maximum range was considered (Fig. 2A). A breakdown of these values for the middle time estimate showed that the scores were distributed differently (Fig. 2B). Option 1 scores high against project duration (ECON 5*), direct economic cost (ECON 1*) and employment capacity (ECON 3), but was offset by the timescale that houses and properties can be released to market (ECON 2 and ECON 4). Option 2 scores lowest against ECON 1* compared with Option 3 and Option 4 due to a lack of treatment plant creating a considerable cost saving. Furthermore, Option 2 only requires a relatively cheap amendment and is not power intensive, although a significant expense comes from a high relative setup cost. This is exemplified by the ratio between the setup and operation and management costs, which are 0.01, 0.53, 0.23 and 0.08, respectively, for Option 1 to 4. These values reflect the fact that while Option 2 does not require treatment plant operation there is a high cost per well due to the initial cost of the electrodes (3×2 m graphite electrodes, ca. £2000 – £2500 per well). Option 4 has a low setup cost as only two wells are predicted to effectively capture the dissolved-phase plume. Conversely, Option 3 requires numerous air sparge wells (predicted radius of influence ca. 3.3 m) to cover the site area, but there is a lower cost per well compared with Option 2.

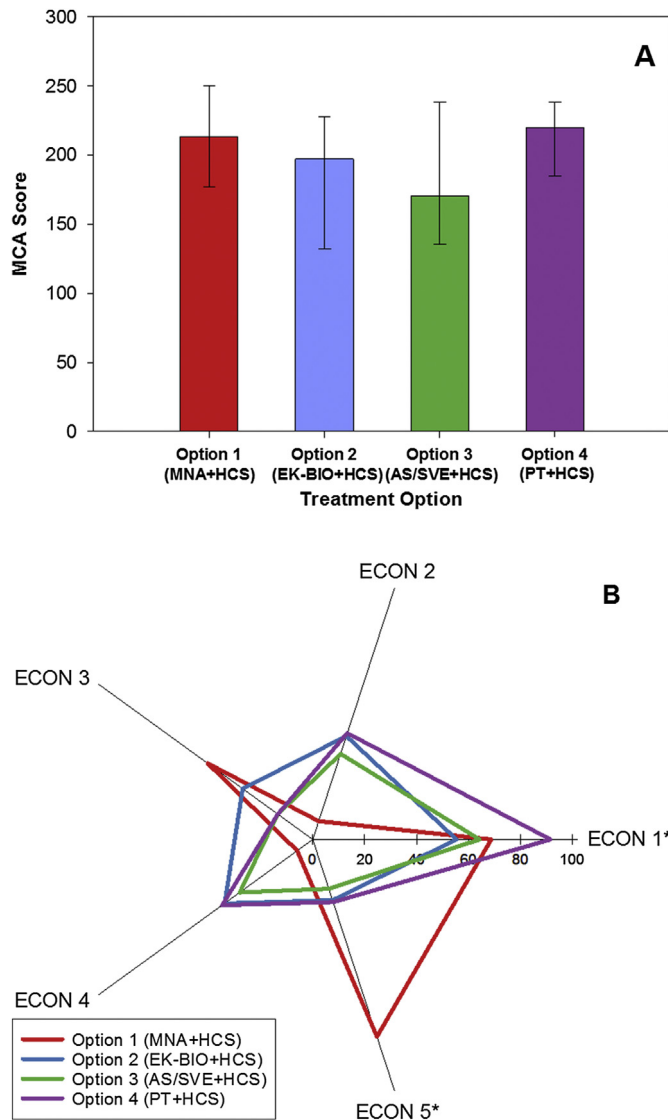


Fig. 2. Results for economic indicators. Two graphs: a bar chart and radar plot. Tier 2 sustainability assessment: Economic ICs. A. is a bar chart showing the cumulative scores for the different ICs. B. is a radar plot that shows the distribution of scores in each IC for the middle estimate scenario, * represents priority ICs. Error bars on A. represent the minimum and maximum estimated values based on uncertainty analysis. For descriptions of ECON 1–5 see Table 4.

Treatment durations for Option 2–4 are within a similar range based on the quantitative uncertainty analysis. They are relatively short compared with Option 1 and so little difference can be identified between them. This influences ECON 2, 3 and 5*, where treatment duration is a key factor.

5.4.2. Environmental indicator categories

Greater difference between options can be identified using environmental ICs (Fig. 3A). Option 1 scores noticeably higher than Option 3 due to a difference between the minimum and maximum MCA scores. This is due to the prolonged impact of Option 1 on soil and groundwater conditions (ENV 2 and ENV 3*, respectively). Conversely, Option 3 has a low associated score due to the benefits of AS-SVE treatment used to improve both soil and groundwater conditions by treating residual LNAPL and extracting VOC from the soil zone.

Option 1 scores highly for ENV 3* which considers the value of

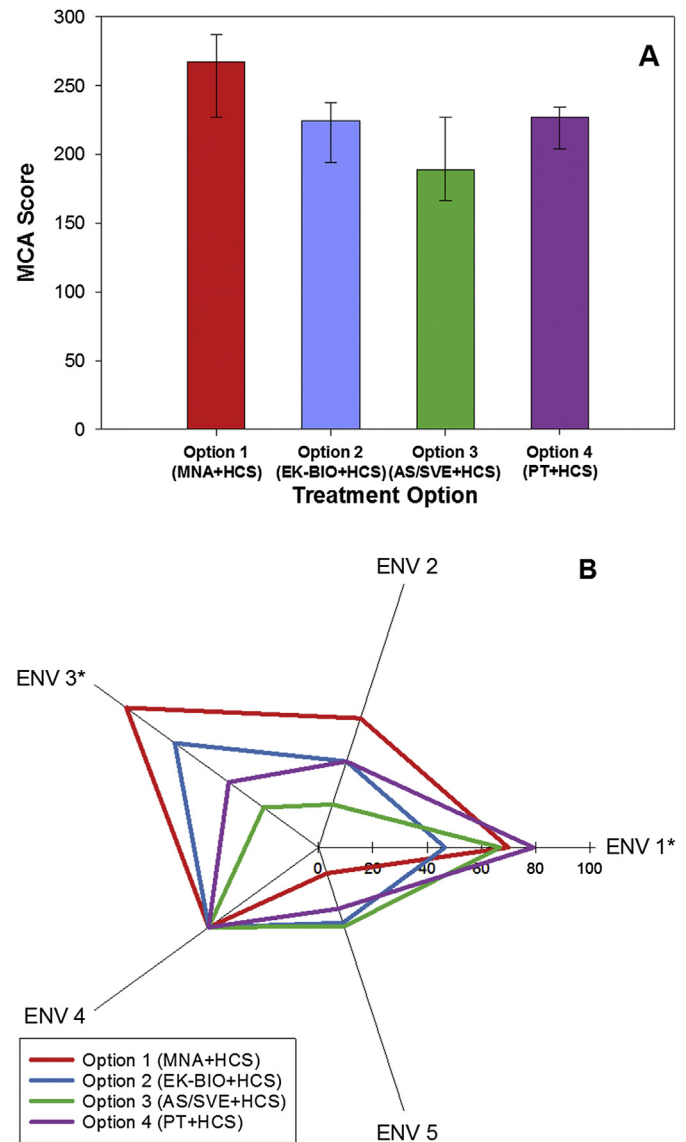


Fig. 3. Results for environmental indicators. Two graphs: a bar chart and radar plot. Tier 2 sustainability analysis: Environmental ICs. A. is a bar chart showing the cumulative scores for the different ICs. B. is a radar plot showing the distribution of scores in each IC for the middle estimate scenario, * represents priority ICs. Error bars on A. represent the minimum and maximum estimated values based on the uncertainty analysis. For descriptions of ENV 1–5 see Table 4.

groundwater abstraction opportunity lost to the water abstracter, based on the volume extracted by the HCS over the treatment period (Bartlett et al., 2014). Option 2 scores high against ENV 3* due to the potential generation of acid and base fronts by uncontrolled electrolysis reactions at the electrodes. However the effect of adding a large mass of sodium nitrate (6–11 t over treatment) is not considered, because it is assumed that the nitrate will be consumed by biodegradation and thus not impact downgradient receptors.

Atmospheric emission of CO₂ (ENV 1*) is driven by several factors, namely treatment power consumption, transport to and from site, and amendment production. Option 2 scores well due to the low electricity requirement; it has an estimated energy demand range between 14 and 30 kW h m⁻³, which compares well to literature values of 19 kWh m⁻³ (Suni et al., 2007). Option 1 and Option 4 have high air emission values due to long-term weekly

visits (road transport) to maintain the HCS and high power requirements for an air stripper and GAC treatment module, respectively.

Option 2 to 4 are not easily distinguishable using the natural resources and waste indicator (ENV 5). ENV 5 combines four categories: water discharged as a result of treatment only (does not include HCS), soil displaced from new well drilling, PVC mass used in new well construction and consumption of fuel. Option 1 scores highly for fuel consumed due to numerous site visits, but low for other values as no new wells are drilled. Option 4 has by far the greatest waste water discharge, compared with other treatments.

5.4.3. Social indicator categories

Against social ICs, Option 1 is best because the maximum MCA score is less than the minimum for the other options (Fig. 4A). This is due to the low level of neighbourhood disturbance associated

with this option, as the MNA treatment requires only periodic visiting and sampling (SOC 3*). Also, the current site investigations and modelling reports for MNA suggest a high level of confidence in the predicted treatment duration, and therefore a low score for the uncertainty and evidence IC (SOC 5). However, a high score is associated with the risk to human health and safety (SOC 1*); this value is calculated using factors for the number of injuries per hour during operations and travel to and from the site. A high score is observed for Option 1 due to the numerous trips to and from site over the treatment period. There is less distinction in the category score between the other options because they have similar treatment duration and there is no safety concern for onsite operations between them if relevant safety procedures are followed. However, Option 2 has the highest level of uncertainty but is predicted to cause less disturbance to local community, due to the absence of a treatment plant (Fig. 4B).

5.4.4. Combined assessment

When the sum of MCA scores for all ICs is considered there is no observable difference between treatments (Fig. 5). For priority ICs, the maximum MCA score for Option 2 is the lowest compared to all other options and overlaps slightly with the minimum score for Option 1 and 4. However, there is an overlap between the minimum and maximum MCA score range for Option 2 and Option 3. Compared against the Tier 1 cumulative rankings for priority ICs, greater differentiation amongst treatments is possible due to the range of uncertainty applied at Tier 2. Similar to the Tier 1 assessment, Option 2 and 3 are indistinguishable; however there is more clarity between Option 1, 2 and 4.

The sensitivity analysis highlights ICs which have the greatest contribution to the MCA score, it is shown in Fig. S5, supporting information. Indirect and induced economic ICs (ECON 2 and ECON 4) are not included for Option 1 because it is assumed that the sale of properties will be after one year and therefore the value is the same between the minimum and maximum treatment durations. When an IC has a low sensitivity it reflects a low range of values relative to other ICs. Thus ICs with the greatest variability will have the greatest effect. The influence of different ICs appears to vary between options. Treatment duration (ECON 5) for example has a greater influence on MCA scores for Option 1 than Options 2–4, whereas air emissions (ENV 1) are less influential than the others.

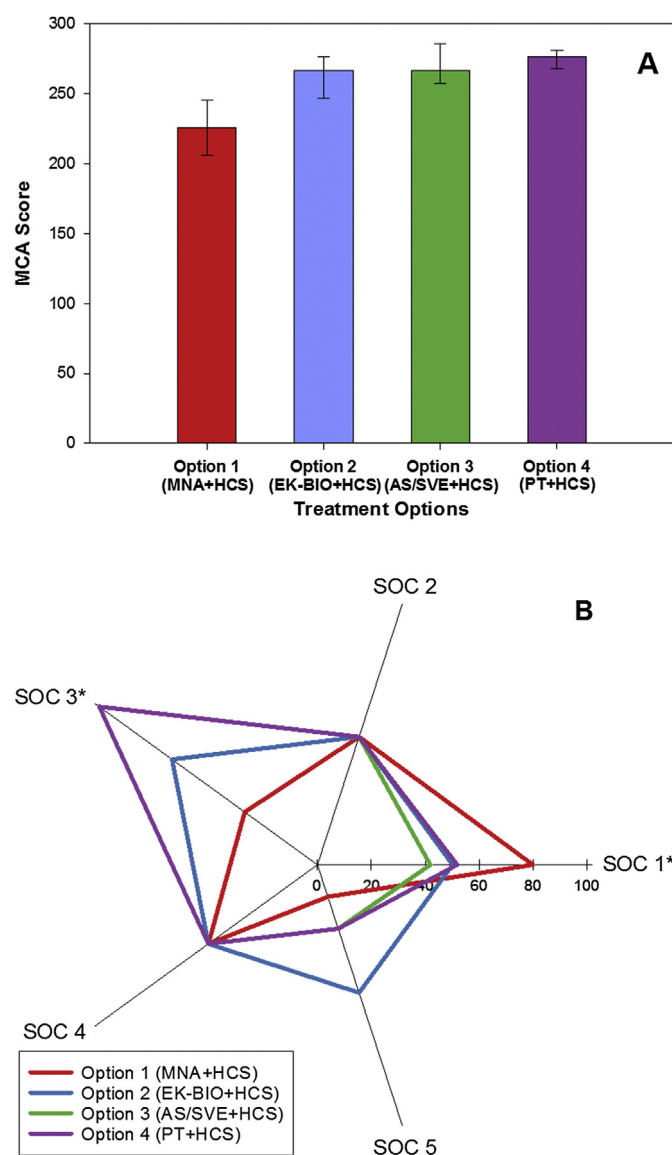


Fig. 4. Results for social indicators. Two graphs: a bar chart and radar plot. Tier 2 sustainability analysis: Social ICs. A. is a bar chart showing the cumulative scores for the different ICs. B. is a radar plot that shows the distribution of scores in each IC for the middle estimate scenario, * represents priority ICs. Error bars on A. represent the minimum and maximum estimated values based on uncertainty analysis. For descriptions of SOC 1–5 see Table 4.

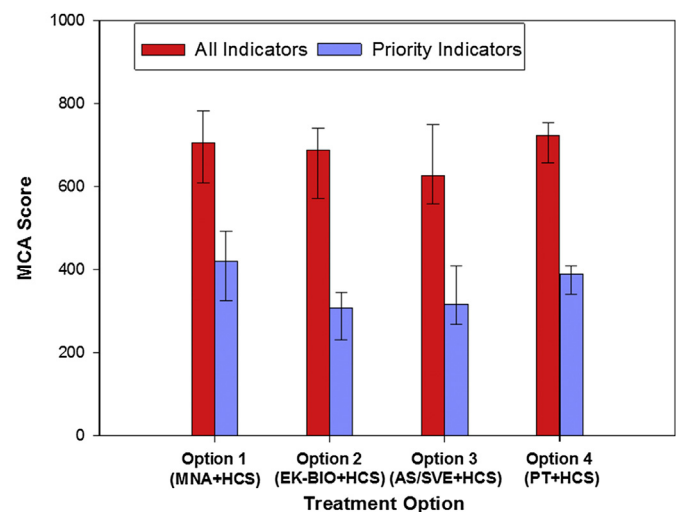


Fig. 5. Bar chart with two data series: 'All ICs' and 'Priority ICs'. Cumulative Tier 2 MCA scores for all and priority ICs. Error bars represent the minimum and maximum estimated values based on uncertainty analysis.

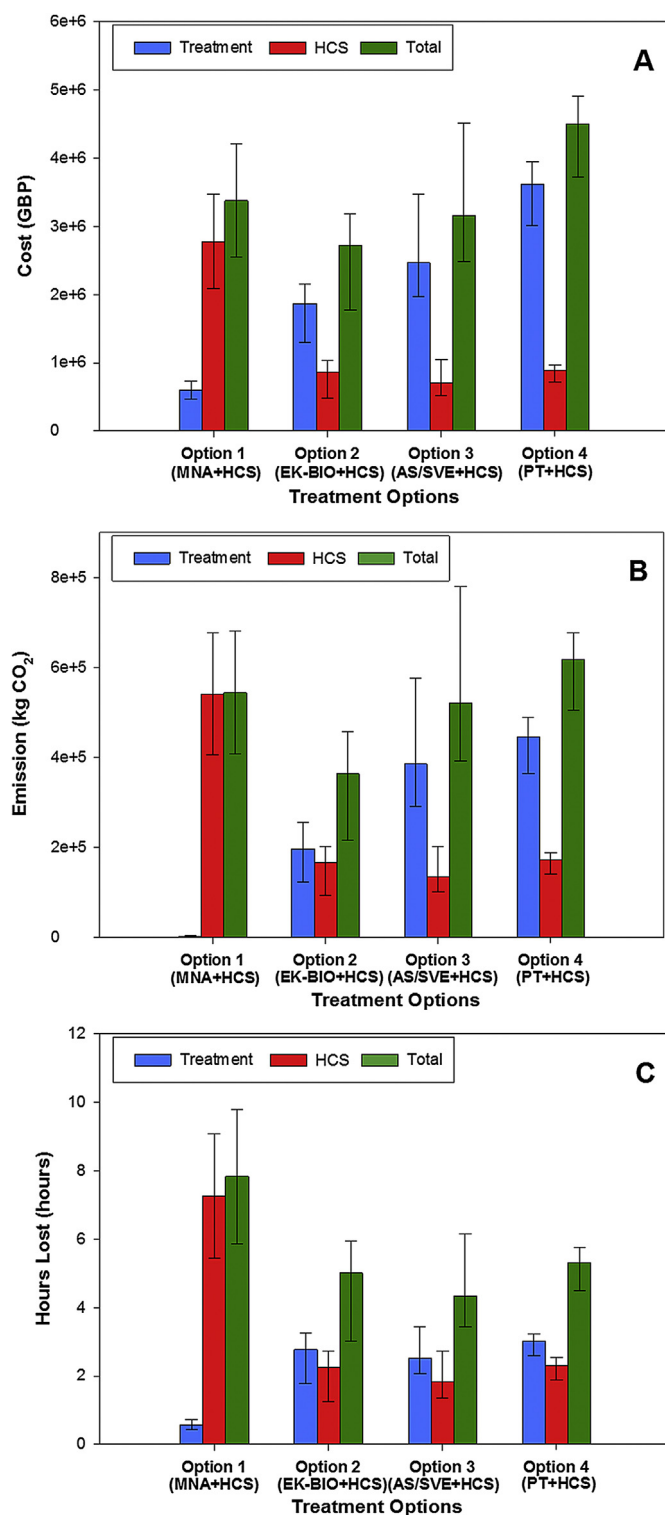


Fig. 6. Three bar charts that describe a breakdown of an individual indicator category. The data series include: 'treatment', 'HCS' and 'Total'. Analysis of individual sustainability ICs. A. ECON 1* total direct cost; B. ENV 1* CO₂ emissions; C. SOC 1* hours lost due to accidents. Error bars represent the minimum and maximum estimated values based on uncertainty analysis.

An analysis of three priority ICs, one from each category, helps identify the role the HCS has in producing the MCA score (Fig. 6A–C). Overall it is clear that for Option 1 the HCS operation is the driver in terms of cost, emissions and health and safety. A value

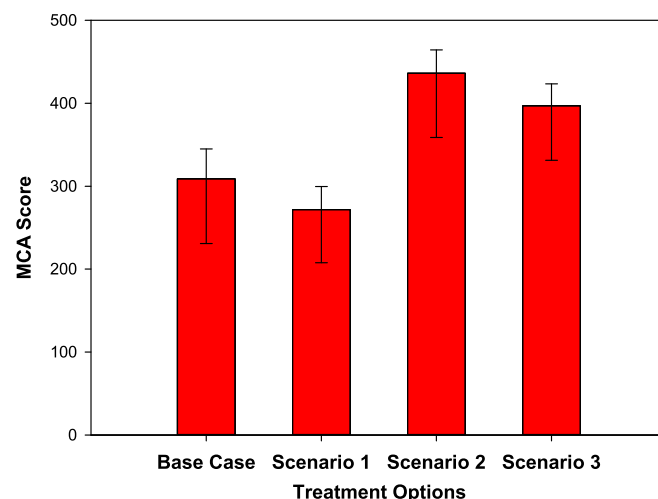


Fig. 7. Bar chart showing priority Indicator category score for different sustainability scenarios. MCA score of priority ICs for four different Option 2 scenarios. Base Case: MCA scores from Tier 2 assessment; Scenario 1: solar panels are main source of electricity; Scenario 2: amendment is replaced with re-circulating groundwater; and Scenario 3: a combination of using solar panels and re-circulating groundwater. Error bars represent the minimum and maximum estimated values based on uncertainty analysis.

for the HCS in other options is similar due to the range of treatment durations. The total direct cost and CO₂ emissions from the treatments increase from Options 2 to 4 and is reflected in the MCA analysis. There is less difference between Options 2 to 4 for health and safety due to similar treatment durations.

5.4.5. Sustainability scenarios

Priority IC scores from MCA analyses of three different treatment scenarios for Option 2 vary significantly depending on the scenario specifications (Fig. 7). Input values for these scores are in Table S19 supporting information. The addition of photovoltaic (PV) panels to Option 2 (scenario 1) does not introduce a significant difference (based on minimum and maximum values) to the base case MCA score (base case and Scenario 1; Scenario 2 and Scenario 3, respectively). The effect is to reduce the total CO₂ emissions and provide a cost saving in operation expenditure, however this is lessened by the need to run pumps consistently and the initial capital expenditure for a PV array approximately 48 m² surface area (see Table S13 supporting information). Conversely, removing the amendment has a significant effect by increasing the MCA score. This is due to an increase in the overall treatment time (e.g. middle estimate rises from 6.2 to 11.3 years) resulting from a decrease in the electron acceptor concentration in electrode wells from amendment to background groundwater supply. As previously highlighted for this CSM, prolonging treatment prolongs HCS operation with subsequent negative effects on sustainability rating. Furthermore, the sodium nitrate amendment is cheap (0.34 GBP kg⁻¹), with a relatively low CO₂ footprint compared with other aspects of the treatment process (e.g. base case middle estimate for amendment production versus transport is 5.1 and 15.8 t, respectively).

6. Discussion

6.1. Tier 1 and Tier 2 assessment outcomes

The SuRF-UK framework is designed to support contaminated land practitioners in identifying the most sustainable remediation

option for a site. In this study, similar results are obtained from both the Tier 1 and Tier 2 assessment; Options 2 and 3 performed similarly well against priority ICs, and better than Options 1 and 4. However, the Tier 2 assessment assigns quantitative values for certain ICs and includes an uncertainty analysis that allows more confidence to be placed in the differences between options. This allows the decision maker to eliminate options relative to the lowest priority IC score, i.e. eliminate Options 1 and 4 based on scores greater than Option 2. Further analysis would be required to differentiate between Options 2 and 3, if it were deemed necessary to show which of these two well-performing options is optimal. Practically, Option 2 would require extensive pilot testing prior to full-scale application to determine whether certain assumptions associated with the electron balance model (e.g. full conversion of nitrate upon contact with contaminated zones) were valid. This data could then be used to inform a Tier 3 sustainability assessment of the two options.

6.2. Comparison of individual treatments

An assessment of remediation options against individual ICs allows the influence of the HCS to be isolated from the treatment (Fig. 6). It is clear that the HCS makes up a greater proportion of the total value compared to the treatment alone for Option 1. Based on the boundaries of the Tier 2 assessment, MNA has the lowest value for CO₂ emissions, total direct cost and hours lost due to remedial activity and thus could be a very sustainable treatment option in the absence of the HCS. This corresponds to a study by Reddy and Chirakkara (2013) who identified MNA as the most sustainable option to treat groundwater for a site contaminated with mixed PAH, heavy metal and pesticides. However, in this example there were no immediate down gradient receptors unlike the present study where the CSM and remedial objectives require the WSW to be isolated from MTBE contamination in the channel sands, hence the need for an active containment system, i.e. the HCS.

Furthermore, out of the intensive treatment options EK-BIO has a lower associated cost and GHG emission footprint. This compares well to an environmental assessment of EK remediation applied to heavy metal contaminated land by Kim et al. (2014). The authors observed during remedial operation that electricity consumption was the highest source of greenhouse gases with little additional extra from transport to and from site. Although overall, the construction stage had the highest associated emissions because the authors included manufacture of consumable materials (e.g. electrodes). The PT option on the other hand has the highest values for total economic cost and CO₂ emissions. Other studies that include PT in the available treatment options often give a poor associated ranking (Cadotte et al., 2007; Reddy and Chirakkara, 2013). This is due to the use of an *ex situ* GAC treatment module and associated manufacture and transport of the GAC to site (Cadotte et al., 2007).

6.3. SuRF-UK framework appraisal

When applying Stage B of the SuRF-UK framework it is important to know the data and time limitations of the project in order to maximise the benefit of the assessment versus the effort expended conducting it. In this study the same management decisions could be reached with a Tier 1 assessment as opposed to a combined Tier 1 and 2 assessment. This is consistent with findings from Smith and Kerrison (2013) where the authors compared the management outcome from Tier 1, 2 and 3 assessments. They found for the same contaminated PFS site that a Tier 2 assessment could differentiate between options ranked equally at Tier 1. Although this did not greatly change the ranking of the options or the final management

decision between assessments. The authors also compared the time and effort required to conduct the assessments and clearly showed it increased between Tier 1, 2 and 3. Overall, the decision to progress between tiers of assessment is highly site and project specific, however it should include consideration of the additional time and data requirements to justify it.

6.4. Sustainability scenarios for EK-BIO

Different Option 2 sustainability scenarios were designed to: (1) offset electricity generation required for the EK-BIO treatment using renewable energy sources, i.e. PVs; and (2) reduce natural resources consumption by replacing the electron acceptor source from the amendment with electron acceptors from background groundwater that is pumped through the electrodes. Both techniques individually (scenarios 1 and 2) and combined (scenario 3) demonstrated limited improvement in the overall sustainability of the treatment. These findings demonstrate that introducing such techniques provide a benefit against green remediation objectives (US EPA, 2008). However, when considered as part of a holistic approach, such as the SuRF-UK framework, these techniques do not always result in a more sustainable solution.

With regards to the PV scenario, there was a small decrease in the score (i.e., improved sustainability), but not significant over the minimum and maximum ranges. Other EK remediation studies have successfully applied PV in their setups at the laboratory- and field-scale (Godschalk and Lageman, 2005; Jeon et al., 2015; Yuan et al., 2009). The noted benefits to using PVs include reduced operational cost and reduced pH changes and corrosion at the electrodes due to the intermittent power supply (Jeon et al., 2015). Furthermore, the remediation efficiency is comparable although slightly lower than systems with a mains direct current supply (Yuan et al., 2009; Jeon et al., 2015). PVs did not significantly reduce the MCA score in this study because: Option 2 is already a comparatively energy efficient option; the HCS contributes half of the CO₂ emissions; and the sensitivity of the influenced IC is relatively low compared to other ICs (air emissions, ENV 1*, Fig. S5, supporting information). Hence, any decrease in cost or CO₂ emissions from incorporating PVs would have a decreased effect on the overall MCA score.

7. Conclusions

This study applies Stage B of the SuRF-UK sustainable remediation framework to inform a management decision for LNAPL and dissolved phase remediation on a complex petroleum fuel-contaminated site. Both a Tier 1 and Tier 2 sustainability assessment are performed using a sustainable indicator set with priority indicator categories selected through a stakeholder workshop. These assessments identified Options 2 and 3 (EK-BIO+HCS and AS/SVE+HCS) as the most sustainable options, whereas Options 1 and 4 (MNA+HCS and PT+HCS) were least sustainable under the conditions of the assessment. However, any application of the EK-BIO option should be subject to pilot-scale testing to ensure assumptions made for the assessment are valid at the field-scale.

In addition to aiding the management decision on site there are wider implications to the study:

- A comparison of individual treatments without the HCS against certain individual ICs highlighted EK-BIO as having lower CO₂ emissions and total cost compared to AS/SVE and PT. This is the first time EK-BIO has been compared to other remediation technologies and shows that it could be a competitive

remediation technology in similar assessments in the future. Moreover, a sensitivity analysis identified which ICs had the greatest contribution to the individual treatment MCA score;

- The effectiveness of the SuRF-UK approach is appraised by comparing the outcome of both the Tier 1 and 2 assessments. Both produced a joint ranking for AS/SVE and EK-BIO treatments, but the uncertainty analysis in Tier 2 gives more confidence in the decision to eliminate the MNA and PT technologies from future analyses, however only proceeding with the Tier 1 assessment could result in time and cost savings; and
- Treatment design modifications intended to make the EK-BIO option more sustainable actually had relatively little effect in changing the sustainability score compared with the other options.

Acknowledgements

This work was completed while the author was in receipt of a UK Engineering and Physical Research Council (EPSRC) Collaborative Awards for Science and Engineering (CASE) (EPSRC grant reference number: EP/J500525/1) studentship with Shell Global Solutions (UK) Ltd.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.jenvman.2016.07.036>.

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